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A study of a membrane bioreactor (MBR) process for landfill leachate treatment

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Wastewater treatment in Finland started in the early 20th century, when the number of water closets increased in the sewerage network of Helsinki and contaminated water caused health and hygiene problems. Landfill leachates have been treated even for a shorter time period, since the legislation regarding them has been inadequate until the 1990s. Over time, tightened legislation has required new innovative solutions from the wastewater treatment in order to meet the stricter requirements for treated wastewater and leachates.

This Bachelor's thesis was done for ArtasFin Oy, a company specialised in designing, manufacturing and installing environmental protection technology such as wastewater treatment plants. The purpose of this case study was to analyse the feasibility of the membrane bioreactor (MBR) process for removing nitrogen, phosphorus and organic matter from landfill leachates in the landfill of Kuopio Waste Centre.

The theory includes background information about the leachate treatment and membrane bioreactor process. Research part includes analysing of the samples taken from the leachates of the landfill of Kuopio Waste Centre, before and after the treatment. In addition, environmental law and regulations in Finland were studied, as well as the conditions and terms of the current environmental permit of Kuopio Waste Centre.

The results and outcomes of the study may be used to design a new wastewater treatment plant for landfill leachates and evaluate the efficiency of the methods used during the piloting period. Further study may be carried out in the future in order to optimize the design of the new wastewater treatment plant even further.

Keywords	MBR, membrane bioreactor, leachate treatment, wastewater treatment

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Jätevesien puhdistus on Suomessa alkoi 1900-luvun alussa WC:iden määrän lisäännyttyä Helsingin viemäriverkostossa ja saastuneen veden jouduttua vesistöihin aiheuttaen terveydellisiä ja hygieenisiä haittoja. Kaatopaikkojen suotovesiä on puhdistettu tätäkin lyhyemmän aikaa, sillä lainsäädäntö on ollut sen suhteen vajavainen aina 1990-luvulle asti. Ajan kuluessa kiristynyt lainsäädäntö on kuitenkin vaatinut jätevedenpuhdistukselta uusia innovatiivisia ratkaisuja, jotta tiukentuneet vaatimukset puhdistetun jäteveden ja suotoveden laadulle täyttyisivät.

Tämä insinöörityö tehtiin ArtasFin Oy:lle, joka on erikoistunut suunnittelmaan, valmistamaan sekä asentamaan ympäristönsuojeluteknologiaa kuten jätevedenpuhdistuslaitoksia. Opinnäytetyön tarkoituksena oli analysoida kalvobioreaktoriprosessin (MBR) soveltuvuutta typen, fosforin ja orgaanisen aineen poistamiseksi kaatopaikkojen suotovesistä Kuopion jätekeskuksen kaatopaikalla.

Kirjallisuusosuudessa tarkasteltiin suotovesien puhdistusta sekä kalvobioreaktoriprosessia. Tutkimusosassa analysoitiin Kuopion jätekeskuksen kaatopaikan suotovesien laatua ennen ja jälkeen jätevedenkäsittelyn. Lisäksi työssä tarkasteltiin Suomen ympäristölainsäädäntöä sekä Kuopion kaatopaikan nykyisen ympäristöluvan ehtoja.

Opinnäytetyön tulokset vahvistavat, että kalvobioreaktoriprosessi soveltuu kaatopaikkojen suotovesien puhdistukseen. Hyviä tuloksia saatiin erityisesti orgaanisen aineen ja typen poistossa. Opinnäytetyön tuloksia voidaan käyttää uuden jätevedenpuhdistamon suunnitteluun suotovesien puhdistuksen osalta ja arvioida pilotissa käytettyjen menetelmien tehokkuutta. Lisätutkimuksia voidaan jatkossa suorittaa, jotta uuden laitoksen mitoittaminen saataisiin suotovesille entistä optimaalisimmaksi.

Avainsanat	MBR, kalvobioreaktori, suotovesien puhdistus, jäteveden puhdistus

Contents

List of Abbreviations

BOD	Biological oxygen demand (mg/l)
COD	Chemical oxygen demand (mg/l)
MBR	Membrane bioreactor
MF	Microfiltration
NF	Nanofiltration
RO	Reverse osmosis
SRT	Solid retention time
UF	Ultrafiltration

1 Introduction

Recent legislation has limited the types of waste that can be disposed in landfills. Currently only a fraction of generated municipal waste ends up in the landfills in Finland. Due to the tightened legislation and regulations, old landfills still require further monitoring and controlling, as they cause both gas and water emissions even after a long period of time. Water emissions occur when water passes through the waste disposed in landfills and different compounds dissolves into it creating leachates. The quality of leachates varies on different landfills. They are affected by, among other things, the characteristics of the waste disposed and the age of the landfill. (Kaartinen, et al., 2009)

In Finland, Environmental Protection Act (527/2014), Environmental Protection Decree (713/2014), Waste Act (646/2011) and the Government Decree on Landfills (331/2013) are governed by the Ministry of the Environment. The purpose of the Government Decree on Landfills (331/2013) is to direct the planning, construction, operation, management, closure and aftercare of landfills in such a way that they will not, even over a long period of time, endanger or cause harm to human health or the environment. (Finlex, 2013) In addition, wastewater treatment is regulated within the EU by the Environmental and Waste Water Directives. However, there are country-specific differences in the directives. In Finland, the activities are monitored by Centre for Economic Development, Transport and the Environment (ELY Centres). Regional state Administrative Agencies are responsible of granting environmental permits. (Elinkeino-, liikenne- ja ympäristökeskus, 2018)

Membrane bioreactor (MBR) is a wastewater treatment process which combines mechanical membrane filtration and biological process, bringing together the benefits of these wastewater treatment processes. This thesis provides information about the treatment of leachates by membrane bioreactors, and it was conducted as literature review together with results from a pilot testing in Kuopio Waste Centre. The environmental permit for the Kuopio Waste Centre has been updated in year 2015, when new limits for the quality of the discharged treated water were given. Artasfin Oy provided a membrane bioreactor pilot plant, a container sized wastewater treatment unit, for Kuopio Waste Centre. The aim of the piloting was to test the suitability of membrane bioreactor for the removal of nitrogen, phosphorus and organic matter from landfill leachate in Kuopio Waste Centre. In addition, the aim of the thesis was to provide solutions and alternative options for the problems encountered during the piloting of the container-sized wastewater treatment unit.

2 Landfills and leachates

Landfilling has been the main disposal method of municipal and industrial waste in Finland until the recent years. The wastewater treatment in Finland started in the early 20th century, when the number of water closets increased in the sewerage network of Helsinki and contaminated water caused health and hygiene problems. However, the regulations and legislations regarding the landfills and leachates were still insufficient until the early 1990s. (Juuti & Rajala, 2017) Due to the lack of legislation, landfills were often established without clarifying the possible environmental risks and without continuous monitoring of the effects on the environment. Even industrial waste and dangerous substances, which are nowadays classified as hazardous waste, have ended up in the municipal waste landfills. Landfills cause stress for the environment and even for human health, as long as the landfilled waste contains substances prone to dissolution, chemical conversion or degradation. (Flöjt, 2010)

2.1 Landfilling in Finland

Finland, among other European countries, is constantly landfilling less waste and finding alternative ways of treating it. In 2016, when the total amount of generated waste was 2768 thousand tons, Finland recycled 42 % of it and exploited 55 % of it as energy, leaving only 3 % of it to landfill. (Tilastokeskus, 2018) The amount of landfilled waste has decreased significantly during the last ten years. More than 50 % of the generated waste ended up in the landfills still in 2008. The development of the amount of municipal waste and the treatment of it can be seen in the Table 1.

Currently, more than half of all generated municipal waste is incinerated in the seven Waste to Energy plants in Finland. The growth of waste incineration in Finland has been rapid, since most of the waste to energy plants have been built after year 2012. (Pöyry, 2015) In addition, the recycling rates have increased in the past years and the target is to increase the recycling of municipal waste so that 55 % of it would be recycled in 2025, 60 % in 2030 and 65 % in 2035. (Ympäristö, 2017)

Table 1. Treatment of municipal waste during the years 1997-2016. (Tilastokeskus, 2018)

	Municipal waste generated (1000 tons)	**Landfill (1000 tons)**	**Landfill (%)**	**Energy (1000 tons)**	**Energy (%)**	**Recycled (1000 tons)**	**Recycled (%)**
1997	2200	1450	65.9	120	5.5	630	28.6
1998	2300	1510	65.7	140	6.1	650	28.3
1999	2400	1480	61.7	200	8.3	720	30.0
2000	2600	1580	60.8	270	10.4	750	28.8
2001	2402	1468	61.1	230	9.6	704	29.3
2002	2384	1485	62.3	216	9.1	684	28.7
2003	2428	1445	59.5	256	10.5	727	29.9
2004	2453	1423	58.0	285	11.6	746	30.4
2005	2506	1478	59.0	227	9.1	801	32.0
2006	2600	1504	57.8	222	8.5	874	33.6
2007	2675	1411	52.7	310	11.6	953	35.6
2008	2768	1406	50.8	478	17.3	884	31.9
2009	2562	1180	46.1	463	18.1	920	35.9
2010	2520	1141	45.3	557	22.1	822	32.6
2011	2718	1093	40.2	678	24.9	947	34.8
2012	2738	901	32.9	925	33.8	912	33.3
2013	2682	672	25.1	1137	42.4	872	32.5
2014	2630	458	17.4	1316	50.0	856	32.5
2015	2738	315	11.5	1312	47.9	1111	40.6
2016	2768	89	3.2	1515	54.7	1164	42.1

The division between landfilling municipal waste, incinerating it into energy and recycling it can be further viewed in the Figure 1 below.

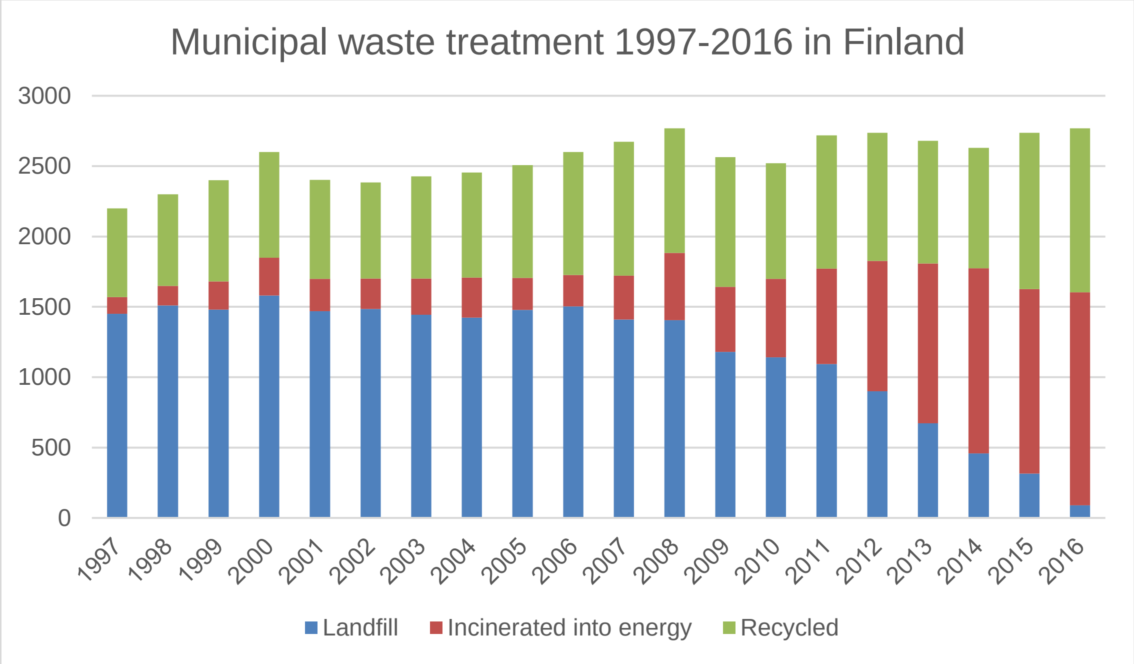

Figure 1. Municipal waste treatment in Finland 1997-2016.

The ambitious recycling targets and increasing incineration of waste to energy will decrease the amount of landfilled waste even further. Although the number of new landfills is dropped to minimum, the old landfills cause environmental risks and hazards as long as there are substances prone to dissolution, chemical conversion or degradation.

2.2 Leachates

Landfill leachates are waters contaminated by wastes in landfills. Leachates are formed when the precipitation, for example, rain or melted snow, runs through the waste dissolving and rinsing different compounds from it. (Kaartinen, et al., 2009)

The quality and quantity of leachate are affected by various factors; the amount and the type of waste, age of the waste, decomposition state of the waste, the size of the landfill, the technique behind the filling of the landfill, water balance of the landfill, permeability of the landfill, inclination of the landfill and the quality of surface waters and climate conditions. In addition, landfill structures can have a significant effect on the formation of leachates. Landfills should be designed so that the precipitation and other waters would not enter the landfill. Wastes should be covered to avoid and minimize the dissolution. Leachate access to the soil shall be prevented by adequate bottom and wall structures. (Kaartinen, et al., 2009)

The internal waters of the landfill are often collected on the bottom of the filling by filter drains. Water is then collected into wells or equalization tanks, from which it can be conducted to a water treatment plant or for a separate treatment. The quality and quantity of the leachates may vary widely, depending on the season. For example, melted snow of one month may account for 25 % of the whole year's precipitation. Equalizing of load peaks with equalization pools or with optimized pumping is often necessary. Depending on the type of waste and the prevailing conditions of the water in landfills, pollutants occurring in leachates are mainly nutrients (nitrogen and phosphorus), salts, suspended solids, dissolved organic matter and metals. (Kaartinen, et al., 2009) The age of the waste has an impact on the organic matter and metals in the leachate. Organic matter concentrations are greater in younger landfills and smaller in older landfills. However, the concentrations of ammonium nitrogen, phosphorus and chloride are not as dependent on the age of waste. Usually, industrial waste landfills include less biodegradable material than municipal waste landfills, and therefore the organic matter and nutrient concentrations are also smaller in them. (Kaartinen, et al., 2009)

2.2.1 Organic compounds

Leachates contain a wide variety of organic compounds, which include easily biodegradable compounds, and not easily biodegradable compounds which require pre-treatment before decomposing due to their strong molecular structure.

BOD (biological oxygen demand) determines the amount of dissolved oxygen required by micro-organisms to break down organic material in the water, whereas COD (chemical oxygen demand) determines the amount of required oxygen to oxidize soluble and particulate organic matter in water. The COD is a measure of total organic carbon content of the water, while BOD measures the easily biodegradable part of the organics. (Kaartinen, et al., 2009)

In Finland, the average BOD content of leachates in young landfills is 2 800 mg/l, whereas the average COD content is 4 600 mg/l. In older landfills the average amounts significantly reduce, BOD being 270 mg/l and COD 550 mg/l. The relationship between BOD/COD indicates the degradation state of waste in the landfill. The smaller the ratio is, the more the waste has degraded. Therefore, the ratio in younger landfills, where the degradation process is still ongoing, is higher than the respective ratio in older landfills.

The BOD/COD ratios is typically 0.4 to 0.5 in younger landfills and 0.1 in older landfills. (Kaartinen, et al., 2009)

2.2.2 Nitrogen

Nitrogen is one of the most significant pollutants in landfill leachates. It does not get stored in waste layers of the landfill, nor is it released into the atmosphere. It is transported from landfills with leachates. The concentrations are high for both young and old landfills, since nitrogen is released from landfill into leachates for decades. (Kaartinen, et al., 2009) Nitrogen is one of the major causes of eutrophication, which means the accumulation of nutrients in waters as a result of human activity. (Ympäristö, 2013)

In leachates, nitrogen is mainly in ammonium nitrogen form (NH_4-N), as a result of the anaerobic degradation of the proteins. Average concentrations of ammonium nitrogen in young landfills in Finland is about 130 mg/l and in older ones 68 mg/l, whereas the total nitrogen content, is about 130 mg/l for younger landfills and 87 mg/l for older ones. Nitrate and nitrite concentrations in leachates are generally very low, since nitrification (ammonium nitrogen oxidation to nitrate) requires oxygen, which is not available in anaerobic conditions of landfill. (Kaartinen, et al., 2009)

2.2.3 Phosphorus

Together with nitrogen, phosphorus is a major cause for eutrophication. (Ympäristö, 2013) Landfill leachates generally contain only small amount of phosphorus compared to sewage waters. Typical total phosphorus content in Finnish landfill leachates is about 2.4 mg/l for young and 0.7 mg/l for older landfills. (Kaartinen, et al., 2009)

2.2.4 Suspended solids

The amount of suspended solids contained in leachate decreases by the age of the landfill. The average solids content of young landfills in Finland is about 127 mg/l and that of old ones is 83 mg/l.

2.2.5 Heavy metals

Metals in leachates include, for example, cadmium, cobalt, copper, chromium, iron, nickel, manganese, lead and zinc. In municipal waste landfills, their concentrations are typically fairly small and even below limit values for drinking water. However, the metal content of leachates may be higher in industrial waste landfills or at landfill sites in waste treatment centres where various types of waste are placed. In younger landfills, the pH is generally low, which makes the dissolvement of metals from waste easier. Older landfills may form aerobic areas where the metals dissolve in water. Typically, metal concentrations in leachates decrease with the age of the landfills. (Kaartinen, et al., 2009)

2.3 Requirements by the law

Landfills and landfill leachates are regulated in Finland by Environmental Protection Act (527/2014), Environmental Protection Decree (713/2014), Waste Act (646/2011) and Government Decree on landfills (331/2013).

Government Decree on landfills (331/2013; amendments until 960/2016 included) regulates planning, establishing, construction, operation, management, direct closure and aftercare of landfills in such a way that they will not, even over a long period of time, endanger or cause harm to human health or the environment. (Finlex, 2013) In addition, it is required to monitor the quantity and quality of landfill leachate and other contaminated water separately at each point where these liquids are conducted away from the landfill site. Also, the treatment of landfill leachate and other contaminated waters and the discharge from treatment shall be monitored to assess the effectiveness of the treatment and the pollution load caused by the landfill reliably. The quality of landfill leachate and other contaminated water shall be analysed at three-month intervals during the operational phase and at six-month intervals during the aftercare phase. (Finlex, 2013)

In addition, Government Decree on landfills (331/2013) requires in section 5, that landfill leachate and other contaminated water shall be collected by means of coordinated subsurface drainage, pumping, or another applicable technical method. The leachate and other contaminated water that has been collected shall be treated effectively at the landfill site or conducted elsewhere for treatment. If the landfill leachate and other contaminated water are conducted elsewhere for treatment, care must be taken that these do not hamper the operation of the wastewater treatment plant or worsen the quality of the sludge generated in it. (Finlex, 2013)

2.4 Requirements by the current environmental permit of Kuopio Waste Centre

Waste management and treatment, and therefore landfills, require an environmental permit by the Environmental Protection Act (527/2014) and the Environmental Protection Decree (713/2014). (Aluehallintovirasto, 2015)

The current environmental permit of Kuopio Waste Centre state that locally treated and slightly polluted leachates which are led to the water courses outside of landfill may contain a total of 35 kg of phosphorus and 400 kg of total nitrogen per year. Organic matter load shall be monitored. (Aluehallintovirasto, 2015)

3 Leachate treatment

Due to the various characteristics of leachates, different treatment methods are required. Treatment of leachate can be biological, physical and chemical. To receive best treatment results, a system which combines more than one treatment method is needed. Coagulation, precipitation, adsorption, membrane processes and some new methods have been added into the leachate treatment procedures to achieve efficient removal of, for example, organic compounds, ammonium, heavy metals and colloidal material. In order to remove the desired chemical compounds, the benefits and weaknesses of various treatment methods should be considered. New advanced methods should be developed together with the development of old conventional methods to keep up with the growing demand of clean water. In Table 2, different treatment options of leachate are listed in order to receive desired results. (Liu, 2013)

Table 2. Treatment options of leachate.

Treatment objectives	Main treatment options
Removal of biodegradable organics (BOD)	Aerobic biological: - Aerated lagoon/extended aeration - Activated sludge - Sequencing batch reactor (SBR) Anaerobic biological: - Upflow sludge blanket
Removal of ammonium	Aerobic nitrification: - Activated sludge - Aerated lagoon/extended aeration - Rotating biological contractor - Sequencing batch reactor - Vegetated ditch (artificial wetlands) Ammonia stripping
Denitrification	Anoxic biological - Activated sludge - Sequencing batch reactor - Vegetated ditch (artificial wetlands)
Removal of non-biodegradable organics and color	Lime / coagulant addition Activated lagoon Reverse osmosis Chemical oxidation
Removal of hazardous trace organics	Activated carbon Reverse osmosis Chemical oxidation
Odor removal	Hydrogen peroxide
Removal of dissolved iron and heavy metals and suspended solids	Lime /coagulant addition, aeration and settling or filtration
Final polishing	Artificial wetlands (e.g. reed beds, ponds)
Disinfection	Hypochlorite
Volume reduction / pre-concentration	Reverse osmosis Evaporation

3.1 Physical/Chemical treatment methods

When the leachate contains a high bio-toxicity, physical/chemical treatment method should be used as a pre- or post-treatment. These methods are used with the biological processes to achieve the required treatment results. Common physical/chemical treatment methods are, for example, coagulation-flocculation, precipitation, flotation, activated carbon adsorption, ammonium stripping, ion exchange, membrane filtration, electro-chemical treatment, chemical oxidation, and advanced oxidation process (AOP). (Liu, 2013)

3.2 Biological treatment methods

Biological treatment includes both aerobic and anaerobic processes and natural systems. In aerobic treatment processes, oxygen is needed for microorganisms to consume organic material for energy and growth. Aerobic treatment methods include activated sludge, sequencing batch reactors (SBR), aerated lagoons, rotating biological contactors, biological aerated filters (BAF) and reed beds. For total nitrogen removal, the biological processes should be designed for nitrification and denitrification. (Liu, 2013)

Unlike the aerobic processes, anaerobic processes take place in the absence of oxygen. Anaerobic treatment methods include, for example, upflow anaerobic sludge blanket (UASB) and anaerobic filters. (Liu, 2013)

3.2.1 Removal of nitrogen

Nitrogen and phosphorus are the main cause of environmental eutrophication. The removal of nitrogen in biological processes is based on nitrification and denitrification. Nitrification converts ammonium nitrogen (NH_4^+ -N) into nitrate (NO_3^+ -N). The nitrifying process is carried out by autotrophic bacteria, which can reduce carbon dioxide to make organic compounds for biosynthesis and receive their energy from an inorganic source. Denitrification process removes nitrate. Denitrification uses heterotrophic bacteria, which are microbes that require organic carbon for growth and as an energy source. They are responsible for the removal of organic carbon compounds in biological treatment processes. (Nissinen, 2014)

3.2.1.1 Nitrification

Nitrification is an aerobic process, which is used to oxidize the ammonium nitrogen sequentially to nitrite and then to nitrate. In the first phase, ammonium nitrogen is oxidized to nitrite according to Equation 1. Nitrite is then oxidized to nitrate according to Equation 2. The overall reaction can be seen in the Equation 3.

$$2NH_4^+ + 3O_2 \rightarrow 2NO_2^- + 2H^+ + 2H_2O \tag{1}$$

$$2NO_2^- + 2O_2 \rightarrow 2NO_3^- \tag{2}$$

Overall reaction:

$$NH_4^+ + 2O_2 \rightarrow NO_3^- + 2H^+ + H_2O \tag{3}$$

Nitrite does not accumulate in most bioreactors since the second step proceeds at faster rate than the first one. Nitrification depends on carbon dioxide, ammonium nitrogen and oxygen. Required dissolved oxygen in the aerobic MBR process is around 1.0-1.5 mg/l (in practice at least 2 mg/l). (Judd & Judd, 2008)

The optimum pH for the nitrification is on average 7.5, and the whole process ceases to function when the pH falls below 6. (Nissinen, 2014) Nitrification consumes alkalinity, and if the wastewater alkalinity is low, pH could be reduced to the levels where nitrification rates are significantly reduced.

Stable or almost complete nitrification is more common in full-scale municipal membrane bioreactor processes than in conventional activated sludge processes. This is most likely due to the smaller floc size in the membrane filtration process, which makes the oxygen transfer easier. However, biomass is a heterogeneous blend in which all the different surface properties affect to the oxygen transfer, for example, through the surface area and the contact surface. Nitrification rate depends on the temperature. This causes significantly decreased efficiency of the removal of ammonium nitrogen when the temperature falls below 10 ° C. (Nissinen, 2014) Figure 2 illustrates the effect of temperature on effluent concentrations of ammonium nitrate (NH_4 -N) and nitrite (NO_2 -N) at different solid retention times.

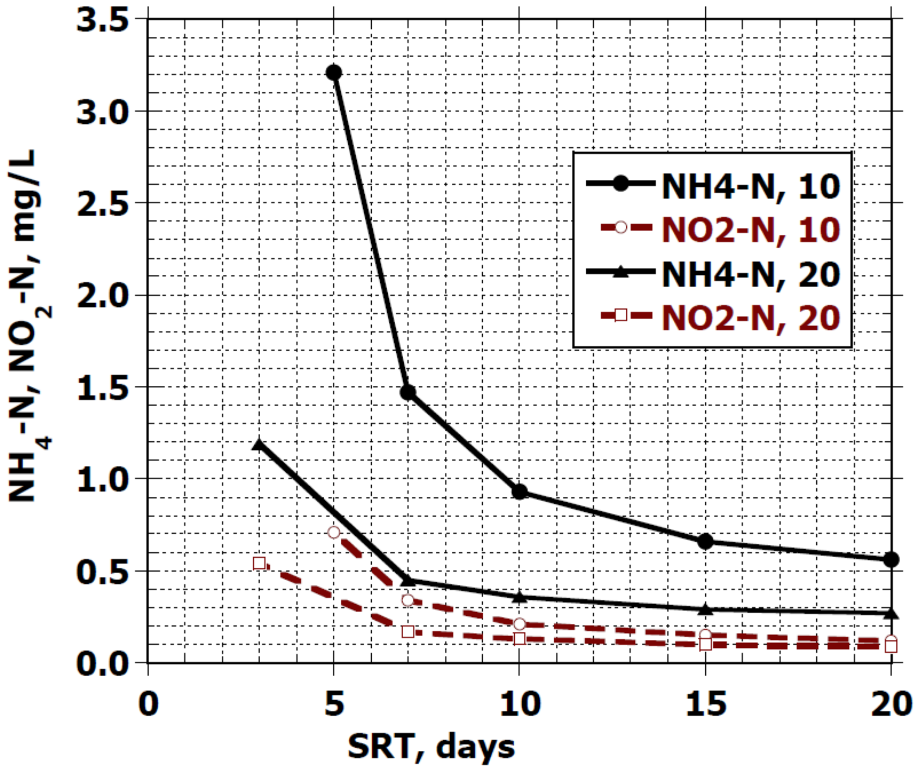

Figure 2. Effect of the solid retention time and temperature of water on ammonium nitrogen and nitrite effluent concentrations. (Nutrient Control Design Manual, 2010)

Longer solid retention time is required at lower temperatures to achieve low concentrations of ammonium nitrogen and nitrite. Nitrite concentrations are also always lower than ammonium nitrogen concentrations. (Nutrient Control Design Manual, 2010)

3.2.1.2 Denitrification

Denitrification takes place under anoxic conditions when oxidation of the organic carbon takes place using the nitrate ion (NO_3) as an electron acceptor, generating nitrogen gas (N_2) as the primary end product:

$$C_{10}H_{19}O_3N + 10NO_3^- \rightarrow 5N_2 + 10CO_2 + 3H_2O + NH_3 + 10OH^-, \qquad (4)$$

where “$C_{10}H_{19}O_3N$” represents organic compounds in wastewater.

Facultative microorganisms, which usually remove organic carbon compounds in an aerobic reaction, convert nitrate into nitrogen gas under anoxic conditions. The denitrification process requires a carbon source for heterotrophic bacteria, which are unable to produce energy without it. The general solution is to recycle the nitrate-rich sludge from aeration and mix it into the incoming wastewater. Full-scale MBR processes are usually designed in such a way that the anoxic process occurs before aeration and membranes. (Nissinen, 2014)

4 Membrane bioreactor process

During the recent years, membrane bioreactor (MBR) technology has become more widespread and accepted as an option for wastewater treatment, replacing conventional activated sludge process. It combines biological activated sludge process and membrane filtration. It is a popular option for many types of wastewaters, whereas the conventional activated sludge (CAS) process cannot cope with either composition of wastewater or fluctuations of wastewater flow rate. (Lignell, et al., 2015) Other advantages of MBR-process include steady quality of water, over 35 % space saving compared to conventional treatment methods, quality of treatment not affected by sludge sedimentation and lower sludge yield. (Lignell, et al., 2015)

The membrane treatment is based on the membranes allowing some physical or chemical components to pass more readily through it than others. Membrane filtration can reject molecular, colloidal and even ion-sized impurities, depending on the pore size of the membranes. Membranes can be divided into four categories based on their pore size: micro, ultra- and nanofiltration and reverse osmosis. (Judd & Judd, 2006) Membranes used in MBRs for treatment of municipal wastewater are in a micro- or ultrafiltration range. The pore size is typically in the range of 0.01 to 0.4 µm, resulting in efficient separation of suspended solids and bacteria. In addition, the separation depends on the material and the porosity of the membrane. (Lignell, et al., 2015)

The pore size of the membranes affects the pressure difference required for filtration. (Lignell, et al., 2015) Figure 3 illustrates the separation efficiencies of different types of membranes. The MBRs typically operate with pressure differences below 0.5 bar, while reverse osmosis may require over 40 bar with sea water desalination.

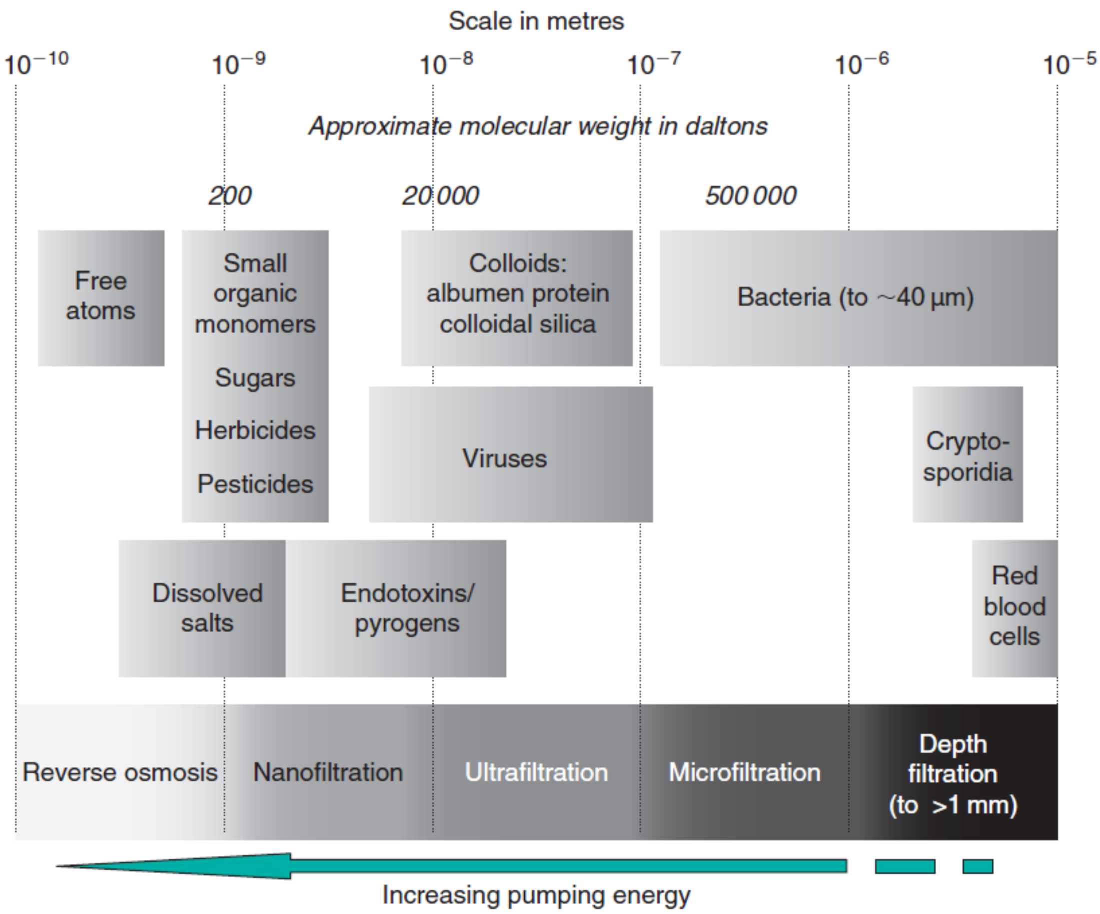

Figure 3. Membrane separation overview (Judd & Judd, 2006)

Ammonium nitrogen, nitrate and nitrite are able to pass through even micro- and ultrafiltration, and therefore the nitrification-denitrification must take place also in the MBR process. (Lignell, et al., 2015) Leachate can also be recycled to landfill to remove nitrogen in the form of nitrogen gas, if the ammonium nitrogen in leachate has been first converted into a nitrate form in a separate treatment process (Rintala, et al., 2001)

4.1 Membrane types and materials

The membrane used in the membrane reactor can be divided into three different configurations: flat sheet, hollow fibre and multi tubular, which are illustrated in the Figure 4. Geometry of the membranes, installation method and their position towards the water flow are important parameters which affect the operation of the treatment process. In addition, it is important to pay attention on how individual membranes are mounted and placed in the membrane modules. Membrane module must be such that the cleaning of the membrane surface by aeration is possible. On the other hand, the membrane modules should have as large filtration surface per unit volume as possible. In MBR process, where membrane modules are immersed directly in the active sludge, flat sheets and hollow fibres are used. Multi tubulars are used in the processes, where the filtration unit is separated from the active sludge. (Lignell, et al., 2015)

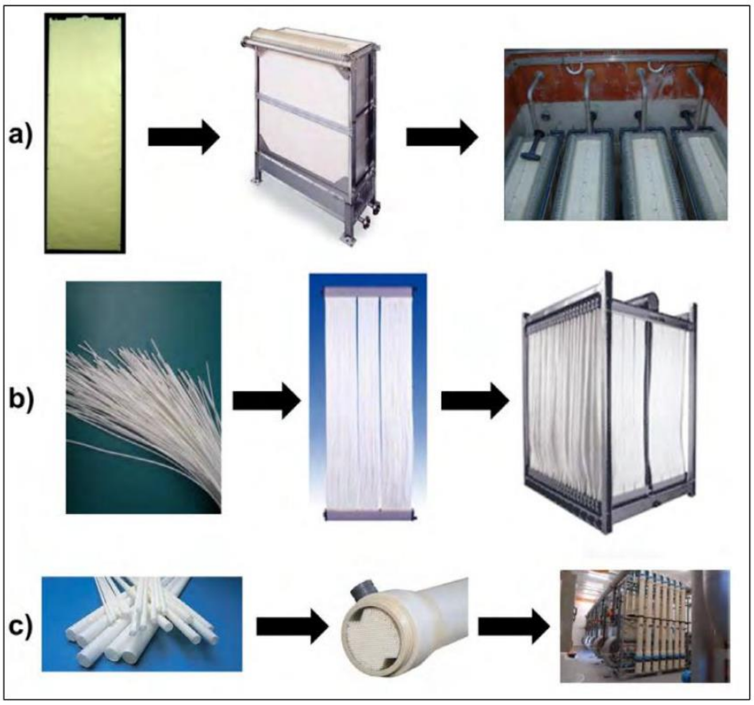

Figure 4. Membranes used in the membrane reactors: a) flat sheet b) hollow fibre and c) multi tubular. (Lignell, et al., 2015)

4.2 MBR processes in landfill leachate treatment

Although MBR technology is still seen as a new technology, the technology has been used successfully for over twenty years around the world, especially in Europe, Asia and North-America. (Melin, 2016) MBR technology has also been applied for landfill leachates. For example, a German company Wehrle Environmental is a MBR process supplier, which specialises in landfill leachate treatment. By 2005, they had installed 58 MBR plants dedicated to leachate treatment. (Judd & Judd, 2008) In 2007, there were between 50 and 60 full-scale MBR installations in operation for treatment of landfill leachate in Germany. (Hai, et al. 2014) The largest MBR plant for landfill leachate treatment is in Bilbao, and it has been in operation since 2004. It is designed to treat a flow of 1.8 million litres per day (MLD), but the highest loads have been measured to be 2.2 MLD. (Judd & Judd, 2008)

In general, landfill leachates are challenging to treat due to high COD loadings and the presence of organics that are not easily biodegradable. (Hai, et al. 2014) In addition, mixed liquor generated from leachate is less filterable than that produced from sewage. Due to this, the energy consumption of MBR-processes is high. (Judd & Judd, 2008)

4.3 Operation and maintenance

The most essential part of the treatment process are the membranes. Therefore, it is important to take care of their maintenance, in order the process to function. Particles accumulate either on the surface of the membranes or in the pores, which will cause decrease in the filtration capacity. This is called fouling. In an event of fouling, the trans-membrane pressure increases and the flux decreases. (Lignell, et al., 2015)

4.3.1 Techniques to control fouling

The most important methods to control fouling are concentration polarization suppression, optimization of physical and chemical cleaning protocols, pre-treatment of the leachate, and mixed-liquor modification. (Radjenovic, J., 2008)

Pre-treatment is important since larger particles accumulate around membrane structures and can even damage the membranes. For MBR, it is recommended to use fine screening of 1-2 mm as a pre-treatment. However, before fine screening, more coarse screens of 3-6 mm can be used. (Lignell, et al., 2015)

The membranes are kept clean by aeration (shear forces induced by air bubbles). Other mechanical cleaning process is a backwash, when the permeate is pumped against the normal filtration direction. (Lignell, et al., 2015) Normally, flat sheet membrane structure does not withstand backwash: therefore it is mainly used in hollow fibre configuration. With flat sheet membranes, relaxation period is used. Relaxation means stopping the filtration for a short period at certain intervals. Sludge age influences the biomass quality, and therefore it will also affect the permeability. By decreasing the sludge age of the process from 10 days to 2 days, fouling of the membranes has been reported to be even 10 times larger. (Lignell, et al., 2015) For membrane fouling control, sufficiently high sludge age should be maintained in the process.

During filtration, material which does not leave by aeration, backwash or relaxation, will accumulate on the membranes. When the trans-membrane pressure has increased to high levels, chemical cleaning should be performed. Chemical washing can be done fully automated or washing can be started manually. Typically, sodium hypochlorite is used to remove organic contaminants and citric acid to remove inorganic contaminants. Instead of hypochlorite, hydrogen peroxide can be used and instead of citric acid, oxalic acid or even hydrochloric acid can be used. Chemical washing procedure varies between membrane types and manufacturers. The cycle of chemical washings varies from daily washings to washings done on a monthly basis. With hollow fibre membranes, chemical washing is performed by adding chemical to the backwash water.

The concentrations used in the chemical cleanings that are performed daily are lower than those used in the maintenance cleanings which are usually performed on a weekly basis. When the fouling cannot be removed by maintenance cleaning, a recovery cleaning is carried out. In the recovery cleaning, the membranes are kept in the cleaning chemical for several hours. For this purpose, the membrane tank must either be emptied of wastewater and filled with chemical, or alternatively, the membrane module must be placed in a separate cleaning tank. The concentrations used in the recovery cleaning are higher than in the maintenance cleaning. Maintenance cleaning usually lasts for about 30 minutes and recovery cleaning for 2-4 hours. The need for chemical cleaning of the flat films is lower, usually 3-4 times a year. (Lignell, et al., 2015)

5 Pilot equipment description

Leachate treatment was tested in Kuopio Waste Centre by a membrane bioreactor pilot plant during autumn 2017. The MBR pilot system is installed in the marine container (Figure 5) and its design capacity is 3 m^3/d.

Figure 5. The MBR-pilot is installed in a marine container.

Figure 6 is a simplified schematic of the pilot process.

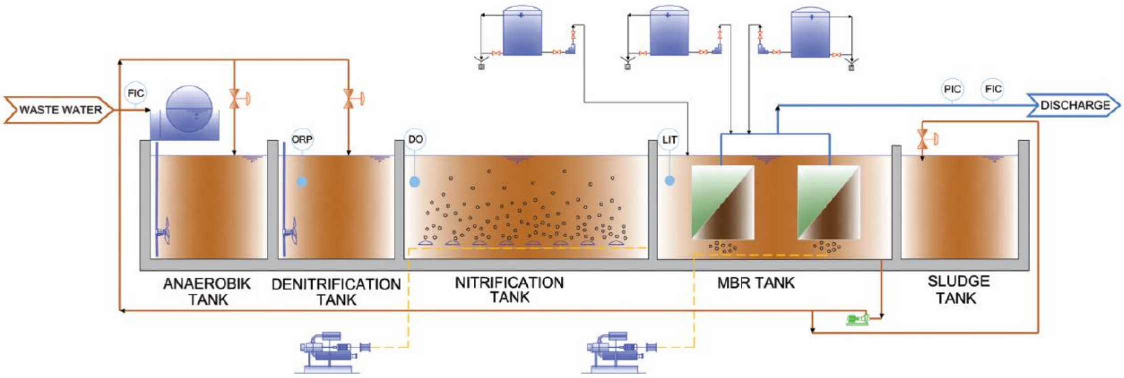

Figure 6. Process diagram of the pilot.

The process is designed for the removal of organic matter (BOD), suspended solids and nitrogen.

The process includes anaerobic tank, denitrification reactor, nitrifying reactor and a membrane reactor (Figure 6). The purpose of the anaerobic tank is to consume the oxygen before denitrification. The oxygen is carried into the anaerobic tank with the sludge recycling. The membranes are kept clean with air bubbles, and therefore the water from the membrane reactor contains dissolved oxygen. Since the pilot is designed for municipal wastewater treatment, denitrification is placed before nitrification in the process. In this case, the organic matter contained in the wastewater is utilized in denitrification. The process requires water recycling, since the nitrate produced in the nitrification should be recycled to the beginning of the process. The next process is the nitrifying reactor, where the nitrifying bacteria oxidize ammonium nitrogen to nitrate. At the same time, the organic material possibly left in the water after the denitrification is biodegraded.

In the membrane reactor, the membranes separate the suspended solids from the water. The pilot has Kubota microfiltration membranes with a pore size of 0.2 µm. The membranes are kept clean by aeration and by stopping the filtration for one minute in nine minutes intervals (relaxation). Biologically treated water is filtered through the membrane by a pump into a treated water tank. Water filtered by the membranes is called permeate. Water collected in the permeate tank is used to dilute the membrane cleaning chemicals.

Landfill leachates contain relatively small amount of organic matter compared to nitrogen. Therefore, additional organic compound (methanol) had to be added to the process for denitrification.

6 Results

The process was started in the beginning of July 2017 by filling the reactors with a sludge taken from the aeration tank of the Kuopio wastewater treatment plant. Continuous test runs and sampling began in September. Three water samples were taken between 25.9.-23.10.2017. During this time, the pressure loss in the membranes had increased and chemical cleaning was required. However, the level sensor in the cleaning system had broken at some point and the process had to be stopped until a new sensor was installed. At the same time, heating cables were installed in water hoses to prevent freezing. The next sampling phase was 10.11-22.11.2017, when one sample was taken before the recycling pump was broken. The recycling was temporarily led into the nitrification tank until the cycle was returned to the beginning of the process by replacing the pumps. After the replacement of pumps, one sample was taken before the process was finally stopped for winter break due to the malfunction of the permeate tank level sensor. The water temperature in the process varied from 11 to 23 °C.

In total, the samples were taken five times during the piloting period of the incoming and treated leachate. The obtained results are described in detail in the following chapters.

pH in raw water and treated water did not differ much, and the recorded values were between 7.9 and 8.3. Nitrification consumes the alkalinity of the water. However, the buffering capacity of the Kuopio leachate is high enough and no separate pH adjustment was required.

6.1 Removal of BOD

On average, there was a 91 % decrease in BOD levels during the piloting period (Table 3).

Table 3. BOD of the incoming and treated leachate.

Date	BOD	
	In (mg/l)	Out (mg/l)
5.10.2017	32	3.0
11.10.2017	20	2.3
19.10.2017	34	2.6
16.11.2017	27	3.3
11.12.2017	33	2.0

Results indicate that the landfill leachate of Kuopio Waste Centre does not contain toxic compounds, at least to the extent that they would prevent microbial activity or that they would be harmful for the environment.

6.2 Removal of COD

On average, there was a 29 % decrease in COD levels during the piloting period (Table 4). This is typical for the leachates of old landfills. The BOD/COD ratio is very low, on average 0.08, which means that only a small fraction of the organic compounds is biodegradable. Most of the easily biodegradable compounds have been depleted from the waste of the landfill and humus type large molecular compounds are left.

Table 4. COD of the incoming and outgoing leachate.

Date	COD	
	In (mg/l)	Out (mg/l)
5.10.2017	350	240
11.10.2017	350	240
19.10.2017	360	250
16.11.2017	350	280
11.12.2017	370	250

If better results for COD removal are desired, an additional process would be required, for example nanofiltration.

6.3 Removal of suspended solids

The removal of suspended solids was good because of the membranes, and almost all of the suspended solids were removed in the process (Table 5).

Table 5. Total suspended solids of the incoming and outgoing leachate.

Date	Suspended solids	
	In (mg/l)	Out (mg/l)
5.10.2017	74	<1
11.10.2017	-	-
19.10.2017	23	<1
16.11.2017	31	2
11.12.2017	27	1.9

6.4 Removal of nitrogen

Nitrification is the most critical part of the biological process because the nitrifying bacteria grow slowly and are sensitive to environmental conditions. Nitrification has mainly worked well and there was a 97 % decrease in ammonium nitrogen levels on average (Table 6). The only exception is the sample taken on 16.11.2017. When the sample was taken, the process most likely had not fully recovered from the break needed for the maintenance.

Table 6. Ammonium nitrogen, nitrite and nitrate of the incoming and outgoing leachate.

Date	Ammonium nitrogen (NH_4-N)		Nitrate (NO_3-N)		Nitrite (NO_2-N)	
	In (mg/l)	Out (mg/l)	In (mg/l)	Out (mg/l)	In (mg/l)	Out (mg/l)
5.10.2017	90	5.6	0.56	49	0.24	0.47
11.10.2017	110	1.9	0.71	55	1	0.034
19.10.2017	110	1.9	0.25	60	0.12	0.059
16.11.2017	120	73	0.28	6.5	0.11	0.4
11.12.2017	130	2.6	0.27	57	0.21	0.09

Nitrate content of the permeate was around 55 mg/l, which was more than what was expected. The methanol dose was probably too low. Methanol input was increased at the end of the piloting period, but the effect was not verified before the experiments had to be stopped. A full scale MBR-equipment includes a nitrogen-measuring sensor which will automatically adjust the methanol dose.

6.5 Removal of total phosphorus

Phosphorus removal in the process was very good, the levels decreased by 97 % on average, although coagulant was not used (Table 7). The phosphorus content of the leachate has been relatively low and bacterial growth has most likely consumed most of the phosphorus. In addition, some of the phosphorus may be in suspended solids which is removed by the membrane process.

Table 7. Phosphorus of the incoming and outgoing leachate.

Date	Phosphorus (P)	
	In (mg/l)	Out (mg/l)
5.10.2017	2.8	0.078
11.10.2017	2.4	0.091
19.10.2017	2.5	0.074
16.11.2017	1.9	0.072
11.12.2017	1.9	0.62

The only exception was the last sample taken on 11.12.2017, where the phosphorus content of the permeate was nearly ten times higher than in the earlier samples. The reason for the deviation of a single sample is unknown. The analysis was performed again, but the concentration was the same; thus, the higher phosphorus content was not an error in the analysis.

7 Conclusions

The aim of the piloting was to test the suitability of membrane bioreactor for the removal of nitrogen, phosphorus and organic matter from landfill leachate in Kuopio Waste Centre. The study was conducted with the analyses taken from leachate before and after the treatment process.

Removal of biodegradable organic matter (BOD) and ammonium nitrogen were good. Removal of BOD was on average 91 %, and removal of ammonium nitrogen was above 98 % in all analyses, except for one. This indicates that the landfill leachate of Kuopio Waste Centre does not contain toxic compounds, at least to the extent that they would prevent microbial activity or that they would be harmful for the environment. The biological process is suitable for the leachate treatment of Kuopio Waste Centre.

The removal of organic matter measured as chemical oxygen demand (COD), was on average 29 %. It is typical for leachates of the old landfills that the biodegradable fraction of organic matter is relatively small. Therefore, COD removal cannot be expected to be as high as in municipal wastewater treatment.

The total nitrogen removal was insufficient because the amount of additional methanol provided for the system was not sufficient for the removal of nitrate (denitrification). A full-scale process should have an on-line sensor for nitrate, which adjusts the methanol dose automatically despite the fluctuations in ammonium nitrogen concentration in the water.

Water temperature is a dominating factor for the nitrification process. The cold climate restrains the sensitivity of total nitrogen, and then affects the nitrification performance. The lowest water temperature was 11 °C, which did not yet affect nitrification in the process.

Adequate phosphorus removal was achieved during the piloting process although no coagulant was added to the process. The amount of phosphorus in the leachate was relatively low and it was removed by the natural phosphorus requirement of the microbes. The removal of phosphorous was on average 91 %. Suspended solids were removed from the water almost entirely by the membranes.

Before the pilot was moved to Kuopio Waste Centre premises, it had not been in operation for two years. This caused some equipment failures which occurred during the piloting period. In addition, increased value of ammonium nitrogen on 16.11.2017 may be a result of pause in the piloting before the analyses.

Since it is required to add additional methanol or other organic matter into the process to remove total nitrogen, it is also possible to locate the denitrification process after nitrification. The process operation would be simplified and oxygen of the recycled water from the membrane reactor can be utilized for nitrification.

Membrane bioreactor process is a suitable choice for leachate treatment when there are strict permit terms of leachate quality and continuously tightening legislation to protect the environment. Since the age of the waste is one of the causing factors of leachate quality, wastewater treatment technologies will have to adapt to the water quality changes in the future

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